1. **Project Name:** **Development of Ultrananocrystalline Diamond Coatings**

2. **Lead Organization:** Argonne National Laboratory
9700 S. Cass Avenue
Argonne IL 60439

3. **Principal Investigator:** John Hryn and John Carlisle
630-252-5894/630-252-1342/hryn@anl.gov
630-252-3520/630-252-9555/carlisle@anl.gov

4. **Project Partners:** Advanced Diamond Technologies (ADT) (in-kind labor)
John Crane, Inc (prototype testing, in-kind labor)
Flowserve (in-kind-labor)
Morgan AM&T (prototype-testing, in-kind labor)
Coorstek Amazing Solutions (in-kind)
Innovative Plasma Systems GmbH (IPLAS) (in-kind)
Industry POC: Neil Kane ADT – 630-953-3507

5. **Date Project Initiated and FY of Effort:** Start Date: Oct 1, 2001, Currently 2nd FY of Effort

6. **Expected Completion Date:** September 30, 2007

7. **Project Technical Milestones and Schedule:** (Please provide the milestones/deliverables schedule for your project, both completed and planned.)

ID Number	Task / Milestone Description	Planned Completion	Actual Completion	Comments
1	**Plasma R&D**			
1.1	6-inch IPLAS rotational holder	9/30/02	6/30/02	constructed 3/31/02
1.2	Optimization of plasma for seals	9/30/02	2/28/02	
1.3	Growth process for seals	9/30/03		75% complete
1.4	Planetary substrate holder (6-in)	9/30/04	3/31/03	
1.5	Seal production proof-of-concept	9/30/04	4/30/03	
1.6	Upgrade IPLAS to 11-inches	9/30/05		Equipment ordered
1.7	Planetary substrate holder (11-in)	9/30/05		
1.8	Optimize 11-inch plasma (single)	9/30/05		
1.9	Optimize 11-inch plasma (multiple)	9/30/06		
1.10	Design fully-automated IPLAS	9/30/06		initiated
1.11	Construct automatic system	6/30/07		
2	**Surface Seeding Study**	**6/30/07**		
2.1	Investigate chlorination of SiC	9/30/02		95% complete
2.2	Develop new seeding process	9/30/03	4/30/03	Optimized for 1” seals
3	**UNCD Material Development**	**6/30/07**		
3.1	Characterization of first 3-in seal	9/30/03	2/28/02	
3.2	Grow UNCD on alpha-SiC seal	3/31/03	3/31/03	Milestone added 12/02
3.3	Grow UNCD on WC seal	6/30/03	4/30/03	Milestone added 12/02

ID Number	Task / Milestone Description	Planned Completion	Actual Completion	Comments
3.4	Grow UNCD on multiple 1" seals simultaneously	9/30/03	4/30/03	Milestone added 12/02
3.5	Grow UNCD films on multiple 3" seals simultaneously with 11" reactor	3/31/04		Milestone added 12/02 Equipment ordered
3.6	Grow UNCD on CDC treated seals	9/30/04	5/15/03	Milestone added 12/02
4	**Pump Seal Application**	**9/30/05**		
4.1	Small Scale Seal Testing	9/30/05		5 tests done
4.2	Installation of Pump Test Loop	9/30/03	3/31/03	Completed by industrial partners
4.3	Long-term Pump Test (6-in IPLAS)	9/30/04		
4.4	Long-term Pump Test (11" IPLAS)	9/30/05		
4.5	Scale-up to proof-of-concept	9/30/05		
5	**Other Applications**	**6/30/07**		**TBD after Task 4**
6	**Final Report**	**9/30/07**		

8. **Past Project Milestones and Accomplishments:** (Provide a brief description of progress and accomplishments to date, with specific emphasis on progress towards milestones during the past calendar year.)

Milestones Achieved over the past year

- New 6" IPLAS Microwave Plasma CVD system installed and commissioned
- Planetary manipulator for 6" reactor installed and commissioned
- UNCD Grown on twelve (12) 3" reaction-bonded SiC seals
- 3" Seals extensively tested in test pump at ANL
- Seeding process/seal pretreatment optimized for 3" reaction-bonded seals
- Successful coating of sintered alpha-SiC 1" automotive seal
- Successful coating of sintered alpha-SiC 2" chemical process pump seal
- Successful coating of 2" WC chemical process pump seal
- Successful coating of multiple (4, 6, & 8) 1" SiC automotive seals simultaneously during one reactor run
    - Seals coated simultaneously passed film uniformity tests at Crane
- Seeding process optimized for 1" SiC seals
- Initial seeding tests performed on UNCD-coated 1" SiC seals
- 12 UNCD-coated 1" SiC seals delivered to Crane for baseline material testing and dynamic testing in test pumps.
- Carbide-derived Carbon (CDC) facility designed
- CDC facility built and commissioned
- 10 CDC coatings formed on 1" alpha-SiC automotive seals
- Initial adhesion testing of CDC coatings on 1" alpha-SiC automotive seals performed
- Broader industrial participant base established
    - John Crane, CoorsTek Amazing Solutions, Morgan AM&T, Advanced Diamond Technologies, Inc., Innovative Plasma Systems GmbH

- Advanced Diamond Technologies, Inc. founded by ANL/UChicago, to commercialize UNCD technology for all applications (Seals, MEMS, sensors, etc.)
- $6.2M proposal written to IMF to scale-up and commercialize UNCD coatings on seals.
- Economic analysis performed by ANL/ADT/Crane, to determine best types of seals to focus on to eliminate technical risks and for most probable applications

9. **Planned Future Milestones:** (Outline your R&D plans and schedule for the remainder of the project, with specific emphasis on plans for the next calendar year.)

Full list of future milestones provided in #7 above. For the upcoming calendar year:

- Critically need new 11" IPLAS reactor to achieve milestones scheduled for 2004.
  - Coating of multiple 2" & 3" seals simultaneously
  - Scale-up and commercialization UNCD deposition tool development
- Plasmas process and seal seeding/pretreatment sensitive to seal material type
  - Carbon nanotube growth on 2" alpha-SiC seals observed (Fe at surface)
  - Phenomenon needs to be resolved for QA of pumps seals
- Once larger proposal gets funded, either through IMF or Chemical Industry IOF, project focus will shift from coating seals to new applications for UNCD materials, specifically:
  - Electrochemical based-sensors (partnership already established with Motorola) to develop sensors for water purification and impurity removal that will have enormous potential for energy savings and environmental benefits in the chemical and forest-products IOF.

10. **Issues/Barriers:** (Provide a brief description of any technical problems or barriers encountered and how these problems have been or will be resolved or significant deviations from original scope and/or budget.)

Primary barrier that has been overcome is the demonstration that UNCD can be taken from basic-science R&D to applied science R&D by establishing a path for commercialization of the technology. A key technical challenge, occasional delamination of UNCD films in severe-duty applications, has been shown to be related to Fe impurities in the SiC seals, and well as the surface seal roughness. Improved seeding processes, including the use of CDC nucleation techniques developed in conjunction with another IMF project, also helped resolve this barrier.

A key issue remains obtaining a larger IPLAS system to determine if the path to commercialization is viable. DOE-BES has agreed to purchase such a unit for UNCD work related to potential MEMS and biosensor applications. The unit will be available for some preliminary work to demonstrate proof-of-concept of uniformly coasting multiple seals in a large reactor. However, several dedicated large IPLAS units must be secured for pilot tests to demonstrate feasible production of UNCD coated seals. A proposal that includes the purchase of large IPLAS units was submitted to the IMF program.

A major barrier identified last year was the need to increase testing capability. This barrier was overcome by increasing the number of industrial participants in the projects and taking advantage of the facilities and expertise of the industrial partners.

11. **Intended Market and Commercialization Plans/Progress:** (Describe the end-use application and market potential for the research, and the plans and progress for commercial application/adoption, where appropriate; be sure to identify what the product of the research will be and how this product will be introduced/disseminated to the appropriate IOFs.)

A company, Advanced Diamond Technologies, was formed by Argonne National Laboratory to be the commercialization vehicle for UNCD. ADT has, over the past few months, developed partnerships and entered into collaborations with leading companies who, like John Crane, will be marketing partners of ADT. In addition to John Crane who is ADT's partner for pump seals, ADT is also working with Intel, Texas Instruments, Delphi Automotive, Motorola and a biomedical device manufacturer. Through ADT, a mechanism exists for rapid, and broad, deployment of UNCD technology to a variety of IOF (and non-IOF) industries.

In the case of pump seals, ADT will be a toll-coating provider to John Crane. It is envisioned that Crane will provide engineered seals to ADT and ADT will add the UNCD coating. With this business arrangement, ADT is free to focus on its competency which is the production of large area UNCD coatings, while leveraging the unmatched marketing and distribution capabilities of Crane worldwide. This arrangement was optimized around speed to market.

ADT has already done the financial modeling to provide justification to the assertion that the UNCD platform can be used to provide cost-effective and market-ready coated seals. Advanced Diamond Technologies was judged "Most Promising Company" at the Nanotechnology Venture Fair in La Jolla, California in September 2002. An R&D 100 award application was submitted.

12. **Patents, publications, presentations:** (Please list number and reference, if applicable.)

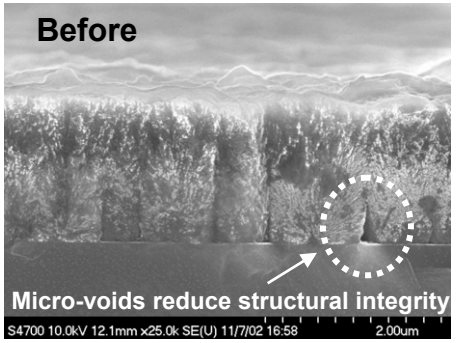
XPS STUDY OF CHEMICAL CONVERSIONS ON SILICON CARBIDE SURFACE TREATED IN CHLORINE-CONTAINING GAS MIXTURES, A.V. Zinovev, J.F. Moore, M.J. Pellin, J. Hryn, O. Auciello, and J.A. Carlisle, to be presented at the Materials Science Forum

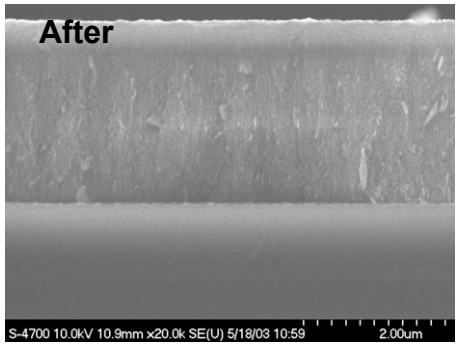
Patents:

"An improved method for seeding carbide substrates for the growth of highly adherent diamond films grown using chemical vapor deposition", J.A. Carlisle, O. Auciello, M. J. Pellin, D.M. Gruen, and J. Hryn, (invention disclosed to Argonne National Laboratory May 23, 2003).

Seal face seeding/pretreatment prevents diamond film delamination

- **We are currently developing diamond coatings (Ultrananocrystalline diamond, UNCD) as wear coatings for mechanical pump seal applications**
- **Delamination of diamond coatings is key failure mode**
- **Work over the past year has focused on surface preparation techniques to eliminate this problem, by improving the mechanical integrity of the interface between the diamond film and the silicon carbon seal, by:**
  - Improved "seeding"
    - *Initial nucleation density has been increased by two orders of magnitude*
  - Surface Pretreatment
    - *Seal surfaced is roughed by mechanical polishing*
- **UNCD-coated mechanical pump seals reduce frictional loss of energy by as much as 20%**
  - 236 trillion Btu savings cumulative by 2020
  - $3.5 billion savings cumulative by 2020





Improved pre-deposition seeding yields increase in initial nucleation density by two orders of magnitude. UNCD/SiC interface now fully dense and mechanically stronger.

Roughening of SiC seal face prior to UNCD deposition has also been shown to reduce film delamination